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Progress in Organic Coatings 55 (2006) 11–19

PROGRESS IN ORGANIC COATINGS

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Film formation from monodisperse acrylic lattices Part 3: Drying and ageing of acrylic latex films

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Received 3 January 2005; accepted 16 September 2005

Abstract

The influence of drying/ageing on the structure and properties of acrylic latex films was investigated using turbidity measurements in combination with gravimetry, scanning electron microscopy (SEM), atomic force microscopy (AFM) and water vapour permeability. Ageing above the minimum film formation temperature (MFFT) leads to marked changes in a dried latex film, whereas, after ageing below the MFFT, changes are hardly found. Above the MFFT there is a continuous change in the film properties with time. This becomes obvious from the decreases in the regenerated interference minimum, water vapour permeability and corrugation height. The influence of ageing on the water absorption of the films is less straight forward. It was expected that films with a more compact structure would absorb less water. This is correct for short drying times only, from 0.5 to 3 h. Ageing of better-dried films, however, yields the opposite result: by increase of the ageing time from 3 to 150 h the water uptake increases. There are various reasons for this increase; they are discussed briefly.

Keywords: Film formation; Monodisperse lattices; Interference; Turbidimetry; Permeability; Atomic force microscopy; Scanning electron microscopy; Water immersion; Water absorption

1. Introduction

The drying process of a latex film usually takes several hours. This time period depends on the characteristics of the dispersions, film thickness and environmental conditions [1]. During a subsequent proper ageing process, morphological changes occur and the film properties (such as mechanical strength and adhesion) are often improved [2–5].

Most industrial lattices contain small amounts of surfactant as a stabilizer [6,7]. During drying and ageing the surfactant may be redistributed in the film and will affect the film properties (e.g. water resistance [8], peel strength and glass transition behaviour [9–11]). On the other hand, some surfactants are compatible enough with latex films and remain distributed evenly throughout the thickness. The redistribution process may start at the very beginning of coalescence and is followed by a slow change with time. After drying, there is a long period of migration towards both sides of the film, due to surfactant incompatibility with polymer, leading to the formation of a parabolic profile of the surfactant concentration through the film thickness [12,13]. During the ageing process, the particle contours could remain [14–17] or vanish [18,19], for example, by migration [3,12]. The surfactant migration is affected by various parameters [3,5–7,12,13,20–27], such as surfactant polymer compatibility and interaction, mobility of surfactant, substrate surface tension, surfactant structure and copolymer composition. The remaining surfactant in a film can absorb moisture and may disrupt the film continuity [28]. The absorbed water in turn (dependent on the sizes of the interstices) can affect the optical properties of the film, such as turbidity. Ordering and coalescence can be studied by measuring transmission spectra (i.e. transmission as a function of wave-length) of latex films during drying. In films prepared from lattices with monodisperse particles, also interference may be present, due to the formation of colloidal crystals. This appears as a more or less sharp minimum in transmission around a certain wave-length, λ_{min}, that primarily is determined by the particle size and concentration, and that in our case lies around 450–500 nm. The state of the prepared film can be studied, by determination of *immersion patterns* of rewetted films (i.e. transmission at a wave-length far

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doi:10.1016/j.porgcoat.2005.09.005

enough beyond the interference minimum, e.g. 600 nm, versus volume fraction of absorbed water in the rewetted film). The water is absorbed in the interstices between the ordered particles and accordingly also a system of ordered water particles of the same "crystal" structure is formed. The background of the method is discussed in references [29–35]. By measuring the transmission spectrum, included the interference of rewetted latex films, information regarding the particle deformation and the amount of absorbed water can be obtained.

In previous papers, we studied the morphology of latex films during colloidal crystallization [36] and the absorption of water in latex films that were dried and subsequently immersed in water [37]. It was illustrated that there are several pitfalls in the determination of water content during immersion, via gravimetrical experiments and via turbidimetry as well [37].

The *first pitfall* is disappearance of boundaries between the particles, so that the before mentioned structure of ordered water particles has changed. In general, we do not have to deal with this pitfall, because from atomic force microscopy (AFM) and scanning electron microscopy (SEM) measurements it appears that it takes a long time before the boundaries between the particles disappear.

The *second pitfall* is the presence of remaining water after insufficient drying. By gravimetric determination of the water content after immersion, this amount of water is overlooked.

The *third pitfall* is a decrease of the particle diameter during the drying process, first a sudden decrease just before the complete disappearance of water in the interstices between the particles, due to the collapse of the protective layer around the particles and the disappearance of the protective electrostatic layer, and secondly a gradual decrease because of a gradual disappearance of water absorbed in the particles themselves.

In the present paper, the influence of drying and ageing on the structure and properties of latex films is investigated using transmission measurements in combination with gravimetry, scanning electron microscopy, atomic force microscopy and water vapour permeability. Methods to get round the mentioned pitfalls are also applied in the present paper.

2. Experimental

2.1. Materials and drying/ageing processes

A monodisperse acrylic latex dispersion (S12) was used (synthesis and characterization were described elsewhere [35,36]). The most important properties are shown in Table 1, where BMA/AA and SDBS mean butyl methacrylate/acrylic acid and sodium dodecyl benzene sulphonate, respectively.

Table 1
Some characteristics of latex S12

Composition	BMA/AA, 97/3 (wt%)
Soap used	SDBS
Solid content	49.4%
MFFT	34 °C
T_g	37 °C
Particle diameter	258 nm
Refractive index polymer	1.485

Thin layers (70 μm) of latex dispersion were brought on an aluminium foil supported by a glass plate and dried in a conditioning chamber during different drying times. Three types of samples were prepared, viz. (a) dried for a short time above the minimum film formation temperature (MFFT), (b) dried and aged for a long time above the MFFT and (c) dried for a short time above the MFFT and subsequently aged below the MFFT. In the short drying time experiments, the latex films were dried at 40 °C and 62% RH during 0.5, 1 and 3 h. In long drying time experiments, the films were dried and aged under the same conditions as the shortly dried films, but now during 3 h, 1 day and 1 week. The third series of samples was prepared by initial drying at 40 °C and 62% RH during 1 h, followed by ageing below the MFFT at room conditions (20 ± 2 °C, 27 ± 5% RH) for 1 month. After preparation of the three series of samples, the thin layers were carefully separated from the aluminium foil and circular samples were cut out with diameters of 2 cm (for transmission, gravimetry and microscopy) and 5 cm (for water vapour permeability). Rewetting by immersion in water was studied during 2–3 weeks. During this process water will be absorbed, and accordingly the light transmission will decrease and the interference minimum will reappear.

2.2. Transmission and interference

During immersion of a dried latex film in water the growing interstitial water inclusions cause turbidity in the rewetted film. This turbidity is characterized by a typical immersion pattern (transmission versus volume fraction of absorbed water in the rewetted film at a certain wave-length), which is calculated with the aid of Rayleigh's transmission law [30,32,35]:

$$T = \exp\left[-\frac{128\pi^5\alpha^2 Nd}{3\lambda^4}\right] \quad (1)$$

where T is the transmission, a the polarizability of the scattering particles (m^3), N the number of scatterers per unit of volume (m^{-3}), d the film thickness (m) and λ is the wave-length of light beam (m).

From Eq. (1) it follows the well-known phenomenon that the transmission decreases with decreasing wave-length.

It was proven by Van Tent and te Nijenhuis [30,31] that the polymer particles are packed according to a hexagonal close packing (hcp) structure, so that the number of scatterers, N, is equal to

$$N = \frac{0.74}{(4/3)\pi r_p^3} \quad (2)$$

where r_p is the radius of the polymer particles.

The number of interstices filled with water is equal to the number of particles as long as the boundaries between the polymer particles has not disappeared. If the water inclusions are small with respect to the particle sizes, then, according to Van Tent and te Nijenhuis [30,31], light scattering can be described quite well by assuming that the water inclusions are ellipsoidal.